ENERGY

Conversion of Hydrogen Sulfide in Coal Gases to Liquid Elemental Sulfur with Monolithic Catalysts

Annual Technical Progress Report for the Period
October 1, 2005 to September 30, 2006

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December 2006

Work Performed Under Contract No
DE-FG26-04NT42129

For
U.S. Department of Energy
National Energy Technology Laboratory
Pittsburgh, PA 15236-0940

By
Tuskegee University
Tuskegee, Alabama 36088

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DISCLAIMER

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SUMMARY

Removal of hydrogen sulfide (H_2S) from coal gasifier gas and sulfur recovery are key steps in the development of Department of Energy's (DOE's) advanced power plants that produce electric power and clean transportation fuels with coal and natural gas. These plants will require highly clean coal gas with H_2S below 1 ppmv and negligible amounts of trace contaminants such as hydrogen chloride, ammonia, alkali, heavy metals, and particulate. The conventional method of sulfur removal and recovery employing amine, Claus, and tail-gas treatment is very expensive. A second generation approach developed under DOE's sponsorship employs hot-gas desulfurization (HGD) using regenerable metal oxide sorbents followed by Direct Sulfur Recovery Process (DSRP). However, this process sequence does not remove trace contaminants and is targeted primarily towards the development of advanced integrated gasification combined cycle (IGCC) plants that produce electricity (not both electricity and transportation fuels).

There is an immediate as well as long-term need for the development of cleanup processes that produce highly clean coal gas for next generation power plants. To this end, a novel process is now under development at several research organizations in which the H_2S in coal gas is directly oxidized to elemental sulfur over a selective catalyst. Such a process is ideally suited for coal gas from commercial gasifiers with a quench system to remove essentially all the trace contaminants except H_2S

In the Single-Step Sulfur Recovery Process (SSRP), the direct oxidation of H_2S to elemental sulfur in the presence of SO_2 is ideally suited for coal gas from commercial gasifiers with a quench system to remove essentially all the trace contaminants except H_2S. This direct oxidation process has the potential to produce a super clean coal gas more economically than both conventional amine-based processes and HGD/DSRP. The H_2 and CO components of syngas appear to behave as inert with respect to sulfur formed at the SSRP conditions. One problem in the SSRP process that needs to be eliminated or minimized is COS formation that may occur due to reaction of CO with sulfur formed from the Claus reaction.

The objectives of this research are to formulate monolithic catalysts for removal of H_2S from coal gases and minimum formation of COS with monolithic catalyst supports, γ-alumina wash or carbon coats, and catalytic metals, to develop a catalytic regeneration method for a deactivated monolithic catalyst, to measure kinetics of both direct oxidation of H_2S to elemental sulfur with SO_2 as an oxidizer and formation of COS in the presence of a simulated coal gas mixture containing H_2, CO, CO_2, and moisture, using a monolithic catalyst reactor, and to develop kinetic rate equations and model the direct oxidation process to assist in the design of large-scale plants. This heterogeneous catalytic reaction has gaseous reactants such as H_2S and SO_2. However, this heterogeneous catalytic reaction has heterogeneous products such as liquid elemental sulfur and steam.

To achieve the above-mentioned objectives using a monolithic catalyst reactor, experiments on conversion of hydrogen sulfide into elemental sulfur and formation of COS were carried out for the space time range of 40 – 560 seconds at 120 – 150^{o}C to evaluate effects of reaction temperatures, total pressure, space time, and catalyst regeneration on conversion of

hydrogen sulfide into elemental sulfur and formation of COS. Simulated coal gas mixtures consist of 3,600 – 4,000-ppmv hydrogen sulfide, 1,800 – 2,000 ppmv sulfur dioxide, 23 – 27 v% hydrogen, 36 – 41 v% CO, 10 – 12 v% CO_2, 0 – 10 vol % moisture, and nitrogen as remainder. Volumetric feed rates of a simulated coal gas mixture to the reactor are 30 – 180 SCCM. The temperature of the reactor is controlled in an oven at 120 – 150^{o}C. The pressure of the reactor is maintained at 40 – 210 psia. The molar ratio of H_2S to SO_2 in the monolithic catalyst reactor is maintained approximately at 2 for all the reaction experiment runs

INTRODUCTION

Coal is our most abundant energy resource. It is strategically important to our nation to increase coal use as an energy source in an environmentally acceptable manner. Coal gasification, a primary step in advanced coal utilization processes, produces a coal gas containing hydrogen (H_2) and carbon monoxide (CO) as the fuel components. Raw coal gas, however, also contains a number of major and trace contaminants including hydrogen sulfide (H_2S), carbonyl sulfide (COS), ammonia (NH_3), hydrogen chloride (HCl), alkali, heavy metals, and particulate. Thus, this gas must be cleaned before further use. H_2S is a major coal gas contaminant that can range from 1000 to 10,000 ppmv, depending on the sulfur content of the coal. Removal of H_2S from coal gas and sulfur recovery are key steps in the development of Department of Energy's (DOE's) advanced Vision 21 plants combining a power plant and a refinery based on coal and natural gas to co-produce electricity and clean transportation-grade liquid fuels. These Vision 21 plants will require highly clean coal gas with H_2S below 1 ppmv and negligible amounts of other contaminants such as COS, HCl, NH_3, alkali, heavy metals, and particulate.

The conventional method of removing H_2S and sulfur recovery involves a number of steps including amine scrubbing at low temperature followed by amine regeneration using steam to produce a concentrated H_2S-containing gas. This concentrated H_2S-containing gas is then combusted to produce a gas with a H_2S to sulfur dioxide (SO_2) ratio of 2 to 1 in a Claus furnace. This is followed by up to three (3) stages of Claus reaction at temperatures of around 250-280^{o}C over an alumina catalyst to recover elemental sulfur:

$$2\ H_2S\ (g) + SO_2\ (g) \leftrightarrow 3S\ (\ell) + 2\ H_2O\ (g)$$

The Claus reaction is exothermic and equilibrium limited. To circumvent equilibrium limitations, the reaction is conducted in up to three (3) reaction stages with interstage cooling/ sulfur condensation followed by interstage re-heating. However, even with three (3) stages, the reaction is not complete due to thermodynamic limitations at 250^{o}C. The Claus tail gas contains sulfur that must be further treated in an expensive tail gas treatment plant (e.g., SCOT) before discharge. Thus, overall H_2S removal and sulfur recovery using this conventional sequence is extremely cumbersome, equipment intensive, and expensive.

A second generation approach for sulfur removal/recovery developed under DOE's sponsorship involves three steps:

(i) hot-gas desulfurization (HGD) using regenerable zinc oxide-based sorbents

$ZnO + H_2S \leftrightarrow ZnS + H_2O$ (HGD)

(ii) sorbent regeneration using air to produce SO_2

$ZnS + {}^3/_2 O_2 \leftrightarrow SO_2 + ZnO$ (regeneration)

(iii) catalytic reduction of SO_2 to elemental sulfur with a small portion of the coal gas in the Direct Sulfur Recovery Process (DSRP):

$SO_2 + 2H_2$ (or 2CO) $\leftrightarrow S + 2H_2O$ (or $2CO_2$) (DSRP)

This approach integrates well with a coal gasifier in an integrated gasification (IGCC) system because the raw coal gas does not have to be cooled all the way down to near room temperature as is the case with the conventional amine/Claus/tail-gas treatment method. However, the overall process scheme requires solid sorbent handling/circulation, and three separate reactors. Also, there is a small energy penalty associated with the use of coal gas to reduce SO_2 by DSRP. Furthermore, since trace contaminants e.g. NH_3 and HCl are not removed by the zinc-based sorbents. This approach is primarily targeted towards the development of advanced IGCC plants that produce electricity only (but do not co-produce both electricity and clean transportation grade fuels).

There is an immediate as well as long-term need for the development of clean processes that produce highly clean coal gas for next generation Vision 21 plants producing both electricity and transportation-grade liquid fuels. To this end, several research organizations are developing a novel process in which the H_2S in coal gas is directly oxidized to elemental sulfur over a selective catalyst using sulfur dioxide (SO_2) produced by burning a portion of the sulfur produced.

The direct oxidation process is ideally suited for coal gas from a commercial gasifier with a quench system. During quench, the trace contaminants (except sulfur) are essentially completely removed and H_2S (with some COS) remains as the only contaminant. The gas contains all of the major coal gas components including H_2, CO, CO_2 and H_2O. Its typical pressure and temperature conditions are 40 to 220 psia and 125 to 155^{o}C. In the direct oxidation process, the Claus reaction is carried out over a selective monolithic catalyst in the presence of the major gas components at around 125 to 155^{o}C to yield liquid sulfur. The low-temperature phase change allows the H_2S-SO_2 reaction to proceed selectively over a catalyst and removes equilibrium limitation. Due to low reactant concentrations, the reaction proceeds nearly isothermally and has the potential to proceed to completion in a single reactor. Burning a required portion of the liquid sulfur in a sulfur burner produces the SO_2 for the process. The process has the potential to produce a super clean coal gas much more economically than both conventional amine-based processes and HGD/DSRP.

The Single-Step Sulfur Recovery Process (SSRP) consists of injecting sulfur dioxide (SO_2) directly into the quenched syngas in the presence of a monolithic catalyst at 125 to 160^{o}C (257 to 320^{o}F) to oxidize H_2S and recover elemental sulfur in a single step via the Claus reaction

($2\ H_2S + SO_2 \rightarrow 3/n\ S_n + 2H_2O$). The SO_2 needed is obtained by burning a portion of the produced sulfur in an external sulfur burner. The key differences between the above-mentioned SSRP process and the traditional Claus process are: (a) in the proposed SSRP process, the Claus reaction occurs in a highly reducing syngas atmosphere containing hydrogen (H_2) and carbon monoxide (CO) and (b) the reaction is carried out at the pressure of the syngas (40-1200 psia). Furthermore, in conventional low-temperature fixed-bed Claus processes e.g. SuperClaus, the catalyst is poisoned by sulfur plugging and must be regenerated by heating externally. In the proposed SSRP process, the liquid elemental sulfur formed from the H_2S removal reaction (Claus reaction) can be detached from the monolithic catalyst surface with the aid of the slip velocity (special flow pattern) of the gaseous reaction mixture at the interface between the thin liquid sulfur layer and the gaseous reaction mixture. The slip velocity, developed from a special flow pattern in monolithic catalyst support channels, induces the pressure difference between the solid catalyst surface and the liquid-sulfur interface by the venturi effect, thereby facilitating catalyst regeneration, sulfur recovery, and favorable shift in thermodynamic limitation on sulfur formation.

Work to date at DOE/NETL, Tuskegee University and RTI has shown the potential of SSRP to convert 99 + % of the H_2S at 40 – 220 psia to elemental sulfur with less than 40 ppmv COS slip. Differential kinetic experiments at Tuskegee University have shown significant increases in rate with pressure increase from 40 to 170 psia. Considering that the commercial SSRP plant will operate at up to 1200 psia, there is potential for complete sulfur removal and recovery. One problem in SSRP that needs to be eliminated or minimized is COS formation that may occur due to reaction of CO with sulfur formed from the Claus reaction.

The objectives of this research are to formulate monolithic catalysts for removal of H_2S from coal gases and minimum formation of COS with monolithic catalyst supports, γ-alumina wash or carbon coats, and catalytic metals, to develop a catalytic regeneration method for a deactivated monolithic catalyst, to measure kinetics of both direct oxidation of H_2S to elemental sulfur with SO_2 as an oxidizer and formation of COS in the presence of a simulated coal gas mixture containing, H_2, CO, CO_2, and moisture, using a monolithic catalyst reactor, and to develop kinetic rate equations and model the direct oxidation process to assist in the design of large-scale plants. This heterogeneous catalytic reaction has gaseous reactants such as H_2S and SO_2. However, this heterogeneous catalytic reaction has heterogeneous products such as liquid elemental sulfur and steam.

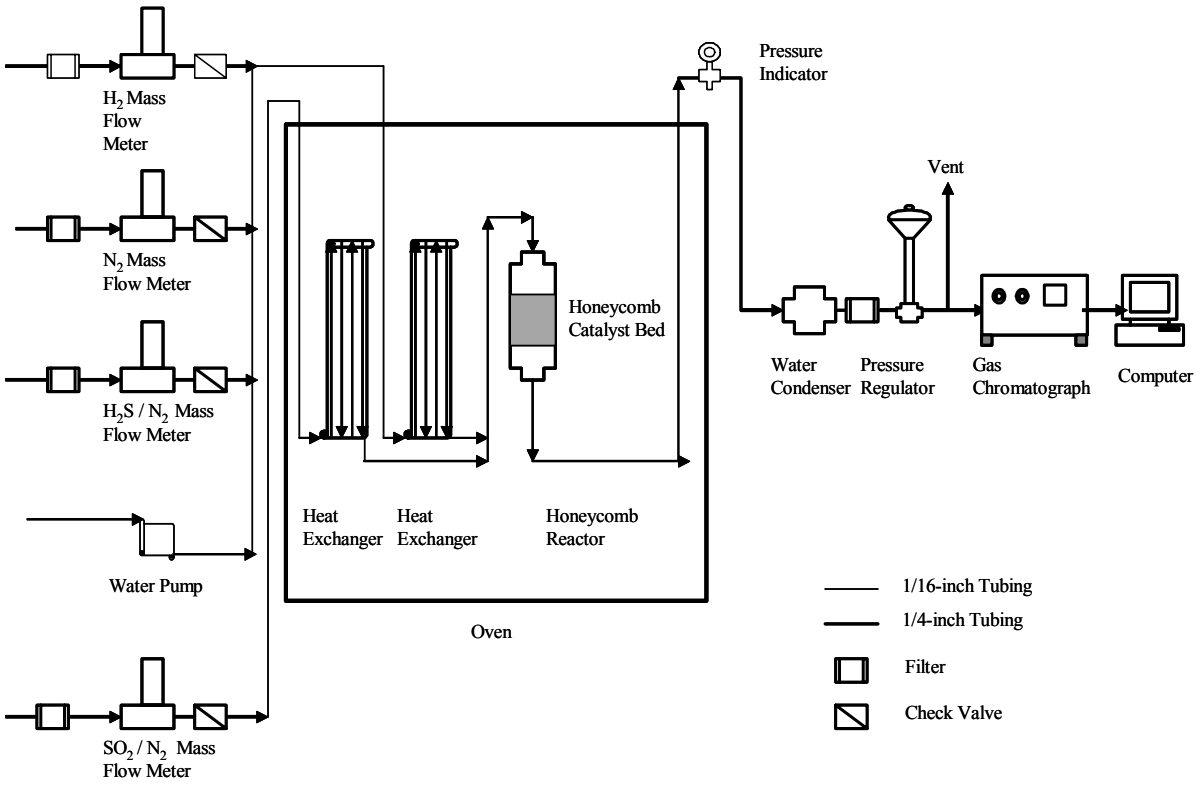
Experiments on conversion of hydrogen sulfide to elemental sulfur and formation of COS using a monolithic catalyst reactor were carried out for the space time range of 46 – 570 seconds at 120 – 150^{o}C and 40 – 210 psia to evaluate effects of space time, reaction temperature, pressure, and catalyst regeneration on conversion of hydrogen sulfide to elemental sulfur and formation of COS. Simulated coal gas mixtures consist of 3,600 – 4,000-ppmv hydrogen sulfide, 1,800 – 2,000 ppmv sulfur dioxide, 23 – 27 v% hydrogen, 36 – 41 v% CO, 10 – 12 v% CO_2, 0 – 10 vol % moisture, and nitrogen as remainder. Volumetric feed rates of a simulated coal gas mixture to the monolithic catalyst reactor are 30 -180 SCCM. The molar ratio of H_2S to SO_2 in the monolithic catalyst reactor is maintained approximately at 2 for all the reaction experiment runs.

EXPERIMENTAL SETUPS

A monolithic catalyst reactor was fabricated with a 2.2-cm inside diameter and 15-cm long 316-stainless steel HPLC column. A γ-alumina wash-coated monolithic catalyst, 2-cm in the diameter and 15-cm long, has 200 square cells and 1040 cm^2 flat surface area. The cell density and the wall thickness of the cordierite monolithic catalyst are 400 square cells/$inch^2$ and 0.02 cm, respectively. A simulated coal gas mixture containing H_2S and SO_2 was reacted with the aid of the catalyst in the monolithic catalyst reactor at 120 – 150°C. Conversion of hydrogen sulfide to elemental sulfur was analyzed with the flame photometric detector (FPD) and the thermal conductivity detector (TCD) of a gas chromatograph. The range of space (residence) time of the reaction gas mixture in the reactor was 46 – 570 seconds under the reaction conditions. Space times are obtained by dividing the bulk volume of the monolithic catalyst in the reactor with the volumetric flow rate of a feed gaseous mixture at reaction conditions.

A reactor assembly mainly consists of four mass flow meters for gases, one reactor, two preheaters, one high pressure liquid pump for water, one four-way switch valve, one oven, five filters for gases, four check valves, and one water collection bottle, as shown in Figure 1. The preheaters are made of 20-ft-long 1/16-inch Teflon tubing.

Figure 1. Schematic Diagram on a monolithic catalyst reactor assembly



The reaction gas mixtures are fed downward to a vertical monolithic catalyst reactor, as shown in Figure 1. The reactor was loaded with a γ-alumina-wash-coated monolithic catalyst in

the vertical reactor. The vertical reactor, loaded with the monolithic catalyst, was placed inside the oven to be heated at a desired temperature. Nitrogen was introduced into the catalyst-loaded reactor during preheating the reactor. When the temperature of the reactor was raised at the desired temperature, one simulated coal gas mixture stream containing H_2S and another feed stream containing SO_2 were introduced into the reactor, by switching nitrogen with the simulated coal gas mixture. The reaction conditions are shown in Table 1. The properties of the monolithic catalyst are shown in Table 2. The experimental data, shown in Tables 3 through 7, were used for drawing the Figures 2 through 12.

Table 1. Experimental conditions for the reaction of hydrogen sulfide with sulfur dioxide as an oxidant using a monolithic catalyst reactor.

Bulk Volume of the honeycomb catalyst bed, cm^3:	47
Temperature, ^oC:	120 -150
Reaction Pressure, psia	40 – 210
Space Time under the reaction conditions, s:	46 – 570
Total Feed Rate, SCCM	30 – 180
Concentration of H_2S, ppmv	3,500 – 4,000
Concentration of SO_2, ppmv	1,800 – 2,000
Carbon Monoxide, v%	36 – 41
Hydrogen, vol %	23 – 27
Carbon Dioxide, vol %	10 – 12
Moisture, vol %:	0 – 10
Nitrogen, vol %	Remainder

Table 2. Dimensions and properties of the monolithic catalyst

Diameter, cm	2
Length, cm	15
Flat Surface Area, cm^2	1,040
Wash Coat	γ-Al_2O_3
Cell Shape	square
Cells/$inch^2$ (CPSI)	400
Wall Thickness, cm	0.02
Flat Area/Length/Cell, cm^2/cm-length/cell	0.345
Chemical Composition	Cordierite ($2MgO\text{-}2AL_2O_3\text{-}5SIO_2$)

Table 3. Conversion of H_2S to elemental sulfur and formation of COS with 30 – 180 SCCM feed streams containing 3,500 – 4,000 ppmv H_2S, 1,800 – 2,000 ppmv SO_2 as an oxidant, 36 – 41 v% CO, 23 – 27 v% H_2, 10 – 12 v% CO_2 and 0 – 10-v% moisture at 120 – 150°C, 40 – 210 psia, and 46 – 570 s space time, regenerating the monolithic catalyst with N_2 at 140°C overnight.

Run	Temp.	Press.	Total Feed Rate, SCCM	Space	Feed Composition, v%							Conversion	COS, ppmv		
#	°C	psia		Time, s	H_2S	SO_2	CO	H_2	H_2O	CO_2	N_2	H_2S	Cylinder Feed	Blank Product Mixture	Reaction Product Mixture
82	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6658	86	46	49
83	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6657	13	54	46
84	140	124	120	143	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5871	5	21	38
85	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5889	17	31	49
86	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5942	5	35	39
87	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6385	6	30	37
88	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6132	5	28	31
89	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.658	5	21	31
90	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.628	5	19	28
91	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6143	8	17	25
92	140	123	120	142	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5833	5	17	25
93	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6248	5	14	23
94	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5808	7	12	26
95	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5556	5	15	31
96	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5322	5	9	31
97	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5108	7	10	30
98	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5283	5	12	31
99	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.4706	6	13	28
100	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3804	6	20	33
101	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.4226	5	26	38
102	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.4367	5	28	37
103	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.46	5	20	34
104	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.4586	6	23	32
105	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.4819	6	24	34
106	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3965	6	25	34
107	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.4122	6	22	32
108	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3687	7	25	32
109	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3467	5	20	31
110	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.1439	5	20	14
111	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.4539	5	16	30
112	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3546	4	16	29
113	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3369	5	16	30
114	140	122	108	157	0.40	0.20	40.76	26.42	0.00	11.21	21.01	0.6944	5	17	37
115	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3791	5	17	33
116	140	124	120	143	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.255	5	17	33
117	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.277	5	17	31
118	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.4745	5	17	57
119	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7071	5	18	48
120	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7043	6	21	37
121	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7316	6	17	34
122	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6234	5	16	32

Table 3. Continued – 1

Run	Temp.	Press.	Total Feed Rate, SCCM	Space	Feed Composition, v%							Conversion	COS, ppmv		
#	°C	psia		Time, s	H_2S	SO_2	CO	H_2	H_2O	CO_2	N_2	H_2S	Cylinder Feed	Blank Product Mixture	Reaction Product Mixture
123	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5652	6	11	33
124	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.666	2	3	3
125	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7365	2	15	36
126	140	123	120	142	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7583	4	16	39
127	140	123	120	142	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7105	4	15	39
128	140	119	120	138	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7196	3	12	33
129	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6919	5	12	37
130	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6659	6	12	37
131	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7311	5	10	41
132	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6198	5	14	43
133	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5458	6	13	38
134	140	119	120	138	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6643	4	14	37
135	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6456	4	14	39
136	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.592	7	15	39
137	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.597	6	11	25
138	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5103	5	11	27
139	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3405	6	15	26
140	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5704	8	12	27
141	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5582	5	14	29
142	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.4586	5	14	30
143	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.4864	5	15	30
144	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3877	5	14	31
145	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3019	5	13	31
146	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3382	5	14	28
147	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3989	6	14	31
148	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3598	6	15	33
149	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3219	5	14	29
150	140	119	120	138	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3677	5	13	28
151	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6641	6	16	48
152	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.8322	6	14	50
153	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.8098	5	14	46
154	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7591	5	17	44
155	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7328	5	16	38
156	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7852	6	17	38
157	120	121	120	147	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7608	6	16	19
158	120	121	120	147	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7479	5	11	17
159	120	123	120	150	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6058	6	16	16
160	120	121	120	147	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7152	5	10	15
161	120	123	120	150	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7584	7	12	18
162	130	122	120	145	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7549	5	12	18
163	130	120	120	142	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7644	5	14	19
164	130	121	120	143	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7445	7	14	20
165	130	121	120	143	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7396	6	12	15
166	150	121	120	137	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7012	5	18	34
167	150	121	120	137	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6718	6	18	32

Table 3. Continued – 2

Run	Temp.	Press.	Total Feed Rate, SCCM	Space	Feed Composition, v%							Conversion	COS, ppmv		
#	°C	psia		Time, s	H_2S	SO_2	CO	H_2	H_2O	CO_2	N_2	H_2S	Cylinder Feed	Blank Product Mixture	Reaction Product Mixture
168	150	121	120	137	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6336	6	17	33
169	140	207	120	239	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7199	6	20	27
170	140	208	120	241	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6891	6	18	27
171	140	209	120	242	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6742	6	17	27
172	140	40	120	46	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.4122	3	15	18
173	140	40	120	46	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3738	8	14	19
174	140	42	120	49	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3088	6	10	11
175	140	41	120	47	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.2483	5	10	11
176	140	41	120	47	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.2544	7	10	11
177	140	119	120	138	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5056	6	12	17
178	140	121	120	140	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.8562	6	23	17
179	140	121	120	140	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.8544	7	14	64
180	140	121	120	140	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.8375	7	14	60
181	140	122	120	141	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.8142	7	14	40
182	140	119	120	138	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.7993	7	14	39
183	140	41	120	47	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.657	7	12	22
184	140	42	120	49	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.614	7	12	19
185	140	40	120	46	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.5954	7	12	19
186	140	40	120	46	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.65	7	11	20
187	140	40	120	46	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.5387	8	13	17
188	140	82	120	95	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6263	8	20	45
189	140	82	120	95	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5387	23	52	48
190	140	210	120	243	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.8553	9	77	79
191	140	209	120	242	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7948	8	83	81
192	140	209	120	242	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.824	7	82	79
193	140	210	120	243	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.8023	9	83	79
194	140	210	120	243	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7943	9	89	84
195	140	172	120	199	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7723	10	81	76
196	140	170	120	197	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7291	9	80	74
197	140	170	120	197	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7162	10	81	73
198	140	171	120	198	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6956	8	86	78
199	140	170	120	197	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6879	11	88	84
200	140	170	120	197	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6212	8	87	81
201	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.8545	8	85	36
202	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7049	9	87	86
203	140	123	120	142	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5886	9	84	78
204	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6938	9	88	81
205	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.8143	8	83	76
206	140	119	120	138	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7235	9	73	77
207	140	120	120	139	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.8134	8	80	73
208	140	119	120	138	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7924	16	79	82
209	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7812	8	73	76
210	140	121	30	560	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.567	9	106	114
211	140	121	30	560	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.2793	8	103	113
212	140	119	30	551	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.8063	8	112	119

Table 3. Continued – 3

Run	Temp.	Press.	Total Feed Rate, SCCM	Space	Feed Composition, v%							Conversion	COS, ppmv		
#	°C	psia		Time, s	H_2S	SO_2	CO	H_2	H_2O	CO_2	N_2	H_2S	Cylinder Feed	Blank Product Mixture	Reaction Product Mixture
213	140	122	30	564	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.8068	8	86	94
214	140	119	30	551	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.634	7	88	91
215	140	119	30	551	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.703	8	97	109
216	140	120	30	555	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7831	9	125	125
217	140	118	30	546	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7292	9	110	113
218	140	117	30	541	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7271	8	102	101
219	140	119	30	551	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6473	9	172	147
220	140	121	60	271	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6674	9	151	143
221	140	117	60	271	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.611	9	97	96
222	140	122	60	282	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6619	8	80	82
223	140	122	60	282	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4787	9	131	129
224	140	120	60	278	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6045	9	94	92
225	140	121	60	280	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6633	7	113	112
226	140	120	60	278	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5664	7	105	107
227	140	119	90	184	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5664	10	95	94
228	140	123	90	190	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4503	8	101	105
229	140	123	90	190	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5049	8	120	122
230	140	120	90	185	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5256	8	149	144
231	140	121	90	187	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4973	8	125	118
232	140	123	90	190	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4222	9	107	110
233	140	124	180	96	0.35	0.18	36.69	23.74	10.00	10.04	19.00	0.3858	9	87	91
234	140	121	180	93	0.35	0.18	36.69	23.74	10.00	10.04	19.00	0.4107	8	104	79
235	140	121	180	93	0.35	0.18	36.69	23.74	10.00	10.04	19.00	0.6036	7	104	85
236	140	120	180	93	0.35	0.18	36.69	23.74	10.00	10.04	19.00	0.5829	7	96	70
237	140	119	180	92	0.35	0.18	36.69	23.74	10.00	10.04	19.00	0.559	7	79	85
238	140	119	180	92	0.35	0.18	36.69	23.74	10.00	10.04	19.00	0.576	8	90	95
239	140	119	120	138	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6186	8	148	131
240	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5767	8	108	108
241	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5133	9	109	111
242	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4976	8	122	116
243	140	123	120	142	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.525	8	101	99
244	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.3917	8	100	95
245	140	124	120	143	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4292	7	99	96
246	140	123	120	142	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4137	8	102	91
247	140	121	120	140	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4717	9	84	82

*SCCM: standard cubic centimeters per minute, volumetric flow rates of gases measured at 1 atm and 25°C

Table 4. Effects of temperature on conversion of H_2S to elemental sulfur and formation of COS with a 120-SCCM feed stream containing 3,600-ppmv H_2S, 1,800-ppmv SO_2, 37-v% CO, 24-v% H_2, 10-v% CO_2 and 10-v% moisture at 120 – 150°C, 120- 123 psia and 137 - 150 s space time, regenerating the monolithic catalyst with N_2 at 140°C overnight.

Run	Temp.	Press.	Total Feed	Space	Feed Composition, v%							Conversion	COS, ppmv		
#	°C	psia	Rate, SCCM	Time, s	H_2S	SO_2	CO	H_2	H_2O	CO_2	N_2	H_2S	Cylinder Feed	Blank Product Mixture	Reaction Product Mixture
151	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6641	6	16	48
152	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.8322	6	14	50
153	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.8098	5	14	46
154	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7591	5	17	44
155	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7328	5	16	38
156	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7852	6	17	38
157	120	121	120	147	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7608	6	16	19
158	120	121	120	147	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7479	5	11	17
159	120	123	120	150	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6058	6	16	16
160	120	121	120	147	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7152	5	10	15
161	120	123	120	150	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7584	7	12	18
162	130	122	120	145	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7549	5	12	18
163	130	120	120	142	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7644	5	14	19
164	130	121	120	143	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7445	7	14	20
165	130	121	120	143	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7396	6	12	15
166	150	121	120	137	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7012	5	18	34
167	150	121	120	137	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6718	6	18	32
168	150	121	120	137	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6336	6	17	33

*SCCM: standard cubic centimeters per minute, volumetric flow rates of gases measured at 1 atm and 25°C

Table 5. Regeneration of the monolithic catalyst for conversion of H_2S to elemental sulfur and formation of COS with a 120 SCCM feed stream containing 3,600-ppmv H_2S, 1,800-ppmv SO_2 as an oxidant, 37-v% CO, 24-v% H_2, 10-v% CO_2 and 10-v% moisture at 140°C, 119 - 123 psia, and 138 – 142 s space time, regenerating the catalyst with N_2 at 140°C overnight.

Run	Temp.	Press.	Total Feed	Space	Feed Composition, v%							Conversion	COS, ppmv		
#	°C	psia	Rate, SCCM	Time, s	H_2S	SO_2	CO	H_2	H_2O	CO_2	N_2	H_2S	Cylinder Feed	Blank Product Mixture	Reaction Product Mixture
118	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.4745	5	17	57
119	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7071	5	18	48
120	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7043	6	21	37
121	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7316	6	17	34
122	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6234	5	16	32
123	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5652	6	11	33
124	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.666	2	3	3
125	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7365	2	15	36
126	140	123	120	142	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7583	4	16	39
127	140	123	120	142	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7105	4	15	39
128	140	119	120	138	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7196	3	12	33

Table 5. Continued – 1

Run	Temp.	Press.	Total Feed	Space	Feed Composition, v%							Conversion	COS, ppmv		
#	°C	psia	Rate, SCCM	Time, s	H_2S	SO_2	CO	H_2	H_2O	CO_2	N_2	H_2S	Cylinder Feed	Blank Product Mixture	Reaction Product Mixture
129	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6919	5	12	37
130	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6659	6	12	37
131	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.7311	5	10	41
132	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6198	5	14	43
133	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5458	6	13	38
134	140	119	120	138	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6643	4	14	37
135	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.6456	4	14	39
136	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.592	7	15	39
137	140	120	120	139	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.597	6	11	25
138	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5103	5	11	27
139	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3405	6	15	26
140	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5704	8	12	27
141	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.5582	5	14	29
142	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.4586	5	14	30
143	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.4864	5	15	30
144	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3877	5	14	31
145	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3019	5	13	31
146	140	122	120	141	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3382	5	14	28
147	140	121	120	140	0.36	0.18	36.69	23.77	10.00	10.09	18.91	0.3989	6	14	31

*SCCM: standard cubic centimeters per minute, volumetric flow rates of gases measured at 1 atm and 25°C

Table 6. Effects of pressure on conversion of H_2S to elemental sulfur and formation of COS with a 120-SCCM feed stream containing 3,500 – 3,600 ppmv H_2S, 1,800-ppmv SO_2, 37-v% CO, 24-v% H_2, 10-v% CO_2 and 10-v% moisture at 140°C, 40 - 210 psia and 46 - 243 s space time, regenerating the monolithic catalyst with N_2 at 140°C overnight.

Run	Temp.	Press.	Total Feed	Space	Feed Composition, v%							Conversion	COS, ppmv		
#	°C	psia	Rate, SCCM	Time, s	H_2S	SO_2	CO	H_2	H_2O	CO_2	N_2	H_2S	Cylinder Feed	Blank Product Mixture	Reaction Product Mixture
178	140	121	120	140	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.8562	6	23	17
179	140	121	120	140	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.8544	7	14	64
180	140	121	120	140	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.8375	7	14	60
181	140	122	120	141	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.8142	7	14	40
182	140	119	120	138	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.7993	7	14	39
183	140	41	120	47	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.657	7	12	22
184	140	42	120	49	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.614	7	12	19
185	140	40	120	46	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.5954	7	12	19
186	140	40	120	46	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.65	7	11	20
187	140	40	120	46	0.36	0.18	36.60	23.72	10.00	10.06	19.08	0.5387	8	13	17
188	140	82	120	95	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6263	8	20	45
189	140	82	120	95	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5387	23	52	48
190	140	210	120	243	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.8553	9	77	79

Table 6. Continued – 1

Run	Temp.	Press.	Total Feed	Space	Feed Composition, v%							Conversion	COS, ppmv		
#	°C	psia	Rate, SCCM	Time, s	H_2S	SO_2	CO	H_2	H_2O	CO_2	N_2	H_2S	Cylinder Feed	Blank Product Mixture	Reaction Product Mixture
191	140	209	120	242	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7948	8	83	81
192	140	209	120	242	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.824	7	82	79
193	140	210	120	243	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.8023	9	83	79
194	140	210	120	243	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7943	9	89	84
195	140	172	120	199	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7723	10	81	76
196	140	170	120	197	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7291	9	80	74
197	140	170	120	197	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7162	10	81	73
198	140	171	120	198	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6956	8	86	78
199	140	170	120	197	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6879	11	88	84
200	140	170	120	197	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6212	8	87	81

*SCCM: standard cubic centimeters per minute, volumetric flow rates of gases measured at 1 atm and 25°C

Table 7. Effects of space time on conversion of H_2S to elemental sulfur and formation of COS with 30 -180 SCCM feed streams containing 3,500-ppmv H_2S, 1,800-ppmv SO_2 as an oxidant, 37-v% CO, 24-v% H_2, 10-v% CO_2 and 10-v% moisture at 140°C, 117 - 124 psia and 90 – 570 s space time, regenerating the monolithic catalyst with N_2 at 140°C overnight.

Run	Temp.	Press.	Total Feed	Space	Feed Composition, v%							Conversion	COS, ppmv		
#	°C	psia	Rate, SCCM	Time, s	H_2S	SO_2	CO	H_2	H_2O	CO_2	N_2	H_2S	Cylinder Feed	Blank Product Mixture	Reaction Product Mixture
201	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.8545	8	85	36
202	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7049	9	87	86
203	140	123	120	142	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5886	9	84	78
204	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6938	9	88	81
205	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.8143	8	83	76
206	140	119	120	138	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7235	9	73	77
207	140	120	120	139	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.8134	8	80	73
208	140	119	120	138	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7924	16	79	82
209	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7812	8	73	76
210	140	121	30	560	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.567	9	106	114
211	140	121	30	560	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.2793	8	103	113
212	140	119	30	551	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.8063	8	112	119
213	140	122	30	564	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.8068	8	86	94
214	140	119	30	551	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.634	7	88	91
215	140	119	30	551	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.703	8	97	109
216	140	120	30	555	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7831	9	125	125
217	140	118	30	546	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7292	9	110	113
218	140	117	30	541	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.7271	8	102	101
219	140	119	30	551	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6473	9	172	147
220	140	121	60	271	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6674	9	151	143
222	140	122	60	282	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6619	8	80	82
223	140	122	60	282	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4787	9	131	129

Table 7. Continued – 1

Run	Temp.	Press.	Total Feed	Space	Feed Composition, v%							Conversion	COS, ppmv		
#	°C	psia	Rate, SCCM	Time, s	H_2S	SO_2	CO	H_2	H_2O	CO_2	N_2	H_2S	Cylinder Feed	Blank Product Mixture	Reaction Product Mixture
224	140	120	60	278	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6045	9	94	92
225	140	121	60	280	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6633	7	113	112
226	140	120	60	278	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5664	7	105	107
227	140	119	90	184	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5664	10	95	94
228	140	123	90	190	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4503	8	101	105
229	140	123	90	190	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5049	8	120	122
230	140	120	90	185	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5256	8	149	144
231	140	121	90	187	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4973	8	125	118
232	140	123	90	190	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4222	9	107	110
233	140	124	180	96	0.35	0.18	36.69	23.74	10.00	10.04	19.00	0.3858	9	87	91
234	140	121	180	93	0.35	0.18	36.69	23.74	10.00	10.04	19.00	0.4107	8	104	79
235	140	121	180	93	0.35	0.18	36.69	23.74	10.00	10.04	19.00	0.6036	7	104	85
236	140	120	180	93	0.35	0.18	36.69	23.74	10.00	10.04	19.00	0.5829	7	96	70
237	140	119	180	92	0.35	0.18	36.69	23.74	10.00	10.04	19.00	0.559	7	79	85
238	140	119	180	92	0.35	0.18	36.69	23.74	10.00	10.04	19.00	0.576	8	90	95
239	140	119	120	138	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.6186	8	148	131
240	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5767	8	108	108
241	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.5133	9	109	111
242	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4976	8	122	116
243	140	123	120	142	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.525	8	101	99
244	140	122	120	141	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.3917	8	100	95
245	140	124	120	143	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4292	7	99	96
246	140	123	120	142	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4137	8	102	91
247	140	121	120	140	0.35	0.18	36.69	23.74	10.00	10.04	19.01	0.4717	9	84	82

*SCCM: standard cubic centimeters per minute, volumetric flow rates of gases measured at 1 atm and 25°C

CALCULATIONS

Concentrations of H_2S and COS in the outlet stream from a monolithic catalyst reactor are analyzed by using a gas chromatograph equipped with a flame photometric detector (FPD) and a thermal conductivity detector (TCD), and a calibration curve. A calibration curve for H_2S is developed with three H_2S samples in different concentrations such as 4,980 ppmv, 996 ppmv, and 249 ppmv, whereas a calibration curve for COS is developed with three COS samples in different concentrations such as 45 ppmv, 30 ppmv, and 15 ppmv.

Each experimental reaction run proceeds after a blank run, which is carried out in the absence of moisture and monolithic catalyst in a reactor. Conversions of H_2S are obtained with concentrations of H_2S from a reaction run and those from its blank run, as shown in the following equation.

$$x = \frac{(C_B - C_R)}{C_B} \quad (1)$$

where x: conversion of H_2S.

C_B: concentration of H_2S in the outlet stream for a blank run.

C_R: concentration of H_2S in the outlet stream for a reaction run

Elemental sulfur is formed with the following reversible stoichiometric reaction formula, as shown in Equation (2). COS is formed in the presence of moisture and catalyst according to the reversible stoichiometric reaction formula, as shown in Equation (3) and Equation (4), which is obtained by adding Equation (2) to Equation (3) multiplied by 3, whereas COS is formed in the absence of moisture and catalyst according to the reversible stoichiometric reaction formula, as shown in Equation (5).

$$2H_2S + SO_2 \leftrightarrow 3S + 2H_2O \quad (2)$$
$$CO + S \leftrightarrow COS \quad (3)$$
$$2H_2S + SO2 + 3CO \leftrightarrow 3COS + 2H_2O \quad (4)$$
$$CO + H_2S \leftrightarrow COS + H_2 \quad (5)$$

RESULTS AND DISCUSSION

Experiments on conversion of hydrogen sulfide into element sulfur were carried out over the space time range of 46 – 570 seconds at 120 - 150^{o}C (see Table 3) to evaluate effects of catalyst age, space time, pressure, and reaction temperature on conversion of hydrogen sulfide into elemental sulfur and formation of COS. Simulated coal gas mixtures consist of 3,600 - 4,000 ppmv hydrogen sulfide, 1,800 - 2,000 ppmv sulfur dioxide, 36 – 41 v% CO, 23 – 27 v% hydrogen, 0 – 12 vol % moisture, 10 – 12 v% CO_2, and nitrogen as remainder. Volumetric feed rates of a simulated coal gas mixture to a monolithic catalyst reactor are 30 - 180 cm^3/min at room temperature and atmospheric pressure (SCCM). The temperature of the reactor is controlled in an oven at 120 - 150^{o}C. The pressure of the reactor is maintained at 40 - 210 psia. Monolithic catalyst is regenerated with nitrogen at 140^{o}C overnight. Multiple experimental data (e.g.: triple, quadruple, quintuple) were obtained and averaged at a given experimental condition to interpret them.

Effects of Temperature on Conversion of H_2S into Elemental Sulfur and formation of COS

Experiments on conversion of hydrogen sulfide to elemental sulfur and formation of COS with a 2-cm-diameter 15-cm-long γ-alumina wash-coated 400-cells/inch^2 monolithic catalyst were carried out over the space time range of 137 - 150 s to evaluate effects of reaction temperature on conversion of hydrogen sulfide to elemental sulfur and formation of COS at 120 - 150^{o}C and 120 – 123 psia. Gas mixtures are fed to a monolithic catalyst reactor containing 3,600-ppmv H_2S, 1,800 ppmv SO_2, 37-v% CO, 24-v% hydrogen, 10-v% moisture, 10-v% CO_2, and nitrogen as remainder. Volumetric feed rates of gas mixtures to the monolithic catalyst reactor are 120 SCCM. Conversion of H_2S to elemental sulfur is 0.60 – 0.84.

Conversion of H_2S to elemental sulfur does not follow the Arrhenius' equation. Reaction temperature affects conversion of H_2S to elemental sulfur. Conversion of H_2S to elemental sulfur increases with increased reaction temperature over the temperature range of 120 -140^oC (see Figure 2), whereas conversion of H_2S to elemental sulfur decreases with increased reaction temperature over the temperature range of 140 -150^oC.

Figure 2. Effects of temperature on conversion of H_2S to elemental sulfur with a 120-SCCM feed stream containing 3,600-ppmv H_2S, 1,800-ppmv SO_2, 37-v% CO, 24-v% H_2, 10-v% CO_2 and 10-v% moisture at 120 – 150^oC, 120- 123 psia and 137 - 150 s space time.

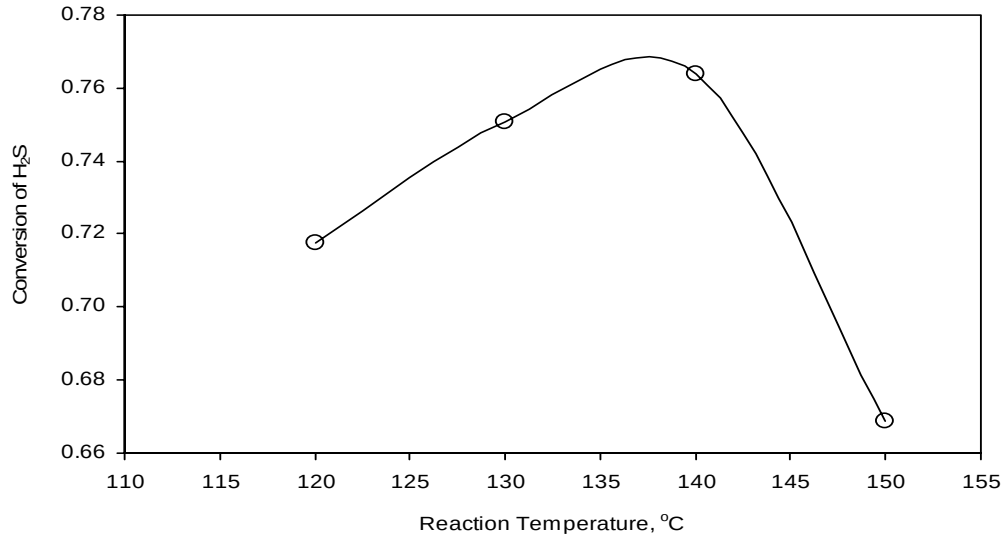


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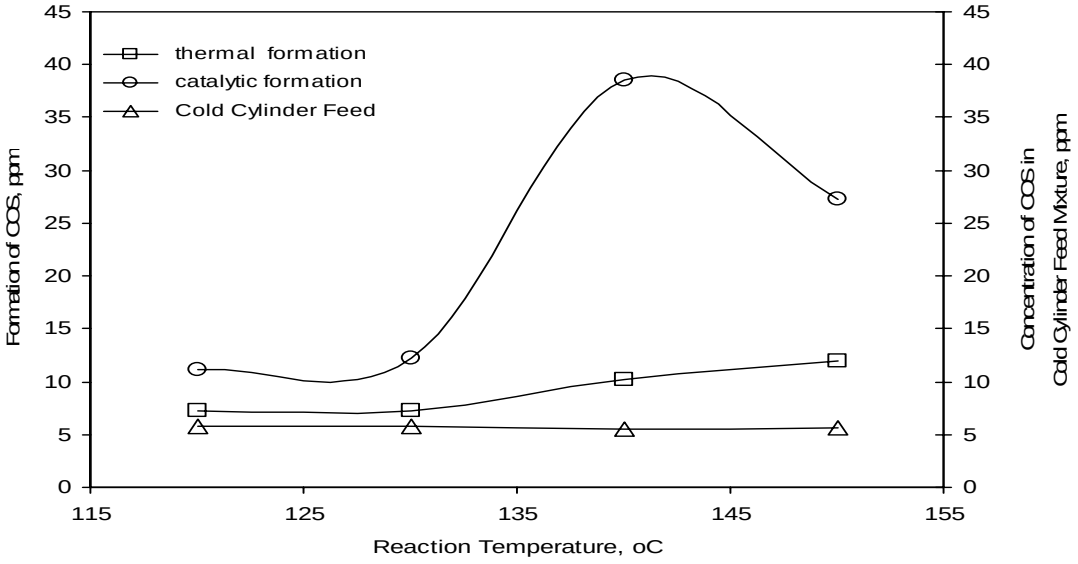
Figure 3. Effects of temperature on formation of COS with a 120-SCCM feed stream containing 3,600-ppmv H_2S, 1,800-ppmv SO_2, 37-v% CO, 24-v% H_2, 10-v% CO_2 and 10-v% moisture at 120 – 150^oC, 120- 123 psia and 137 - 150 s space time.



Catalytic formation of COS levels off and is minimum over the temperature range of 120 -130°C, increases with increased reaction temperature over the temperature range of 130 -140°C, and decreases with increased reaction temperature over the temperature range of 140 -150°C, as shown in Figure 3. Thermal formation of COS levels off over the temperature range of 120 - 130°C, and increases with increased reaction temperature over the temperature range of 130 - 150°C. Catalytic formation of COS is higher than thermal formation of COS.

Figure 4. Effects of conversion of H_2S on formation of COS with a 120-SCCM feed stream containing 3,600-ppmv H_2S, 1,800-ppmv SO_2, 37-v% CO, 24-v% H_2, 10-v% CO_2 and 10-v% moisture at 120 – 150°C, 120- 123 psia and 137 - 150 s space time.

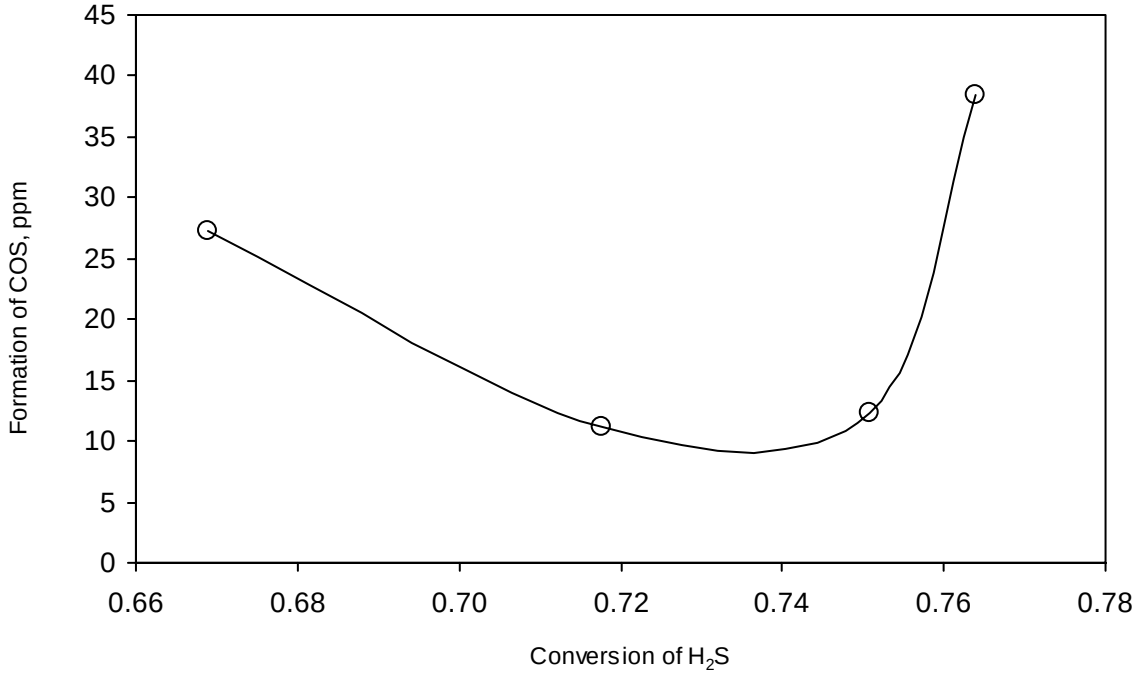


Figure 4 indicates that catalytic formation of COS is minimum over the H_2S conversion range of 0.72 – 0.75 and also over the temperature range of 120 – 130°C.

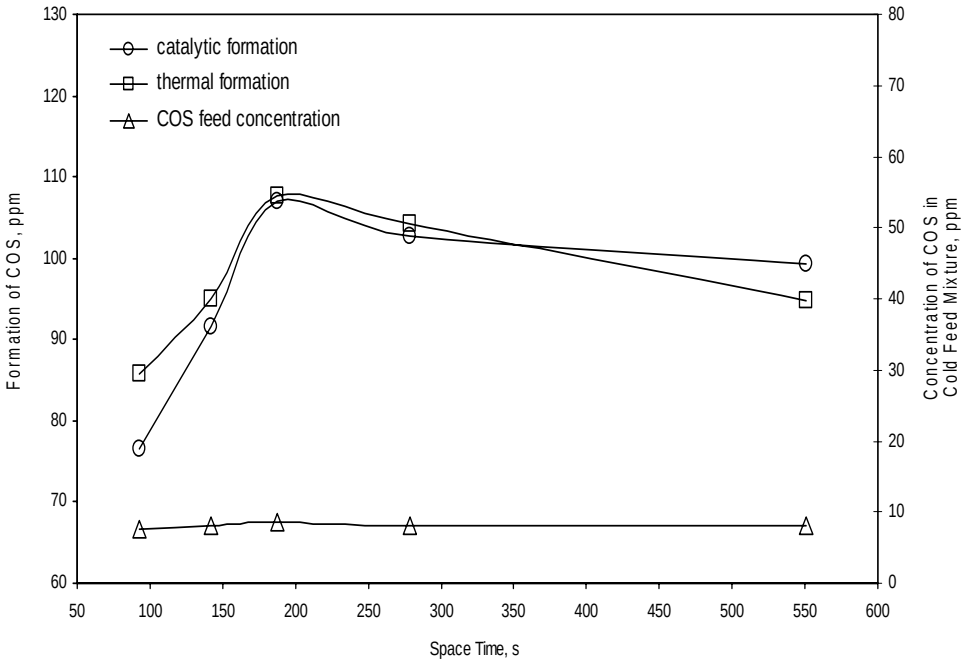
Effects of Space Time on Conversion of H_2S into Elemental Sulfur and formation of COS

Experiments on conversion of hydrogen sulfide to elemental sulfur with a 2-cm-diameter 15-cm-long γ-alumina wash-coated 400-cells/inch^2 monolithic catalyst were carried out over the space time range of 90 – 570 s, which is developed by increasing total volumetric flow rate of gaseous feed mixture to a monolithic catalyst reactor from 30 SCCM to 180 SCCM to evaluate effects of space time on both conversion of hydrogen sulfide to elemental sulfur and formation of COS at 140°C and 117 -124 psia. A gas mixture fed to the monolithic catalyst reactor contains 3,500-ppmv H_2S, 1,800-ppmv SO_2, 37-v% CO, 24-v% hydrogen, 10-v% moisture, 10-v% CO_2, and nitrogen as remainder.

Conversion of H_2S to elemental sulfur is 0.27 – 0.86. Concentration of COS in the outlet stream from the catalyst-loaded reactor is 36 – 147 ppmv, whereas concentration of COS in the

outlet stream from the reactor in absence of the catalyst and moisture is 73 – 172 ppmv. Space time affects conversion of H_2S to elemental sulfur, and formation of COS for both the reaction runs and the blank runs over the pressure range of 117 -124 psia, as shown in Figures 5 and 6.

Figure 5. Effects of space time on formation of COS with 30 - 180 SCCM feed streams containing 3500-ppmv H_2S, 1800-ppmv SO_2, 37-v% CO, 10-v% CO_2, 24-v% H_2 and 10-v% moisture at 140^oC, and 117 - 124 psia and 90 – 570 s space time.



Conversion of H_2S to elemental sulfur increases with space time, as shown in Figure 6. Formation of COS for both the reaction runs and the blank runs increases with space time over the space time range of 90 – 190 s and decreases with increased space time over the space time range of 190 – 550 s. Catalytic formation of COS for the reaction runs is slightly lower than thermal formation of COS for the blank runs over the space time range of 140 – 280 s. Catalytic formation of COS for the reaction runs is much lower than thermal formation for the blank runs at the space time 90 s, whereas catalytic formation of COS for the reaction runs is much higher than thermal formation of COS for the blank runs at the space time 550 s.

Catalytic formation of COS for the reaction runs appears to be independent of conversion of H_2S to elemental sulfur over the space time range of 90 – 570 s, as shown in Figure 7. These facts may suggest that COS be formed by reacting H_2S with CO ($CO + H_2S \rightarrow COS + H_2$)

Figure 6. Effects of space time on conversion of H_2S with 30 - 180 SCCM feed streams containing 3500-ppmv H_2S, 1800-ppmv SO_2, 37-v% CO, 10-v% CO_2, 24-v% H_2 and 10-v% moisture at 140°C, 117 -124 psia and 90 – 570 s space time.

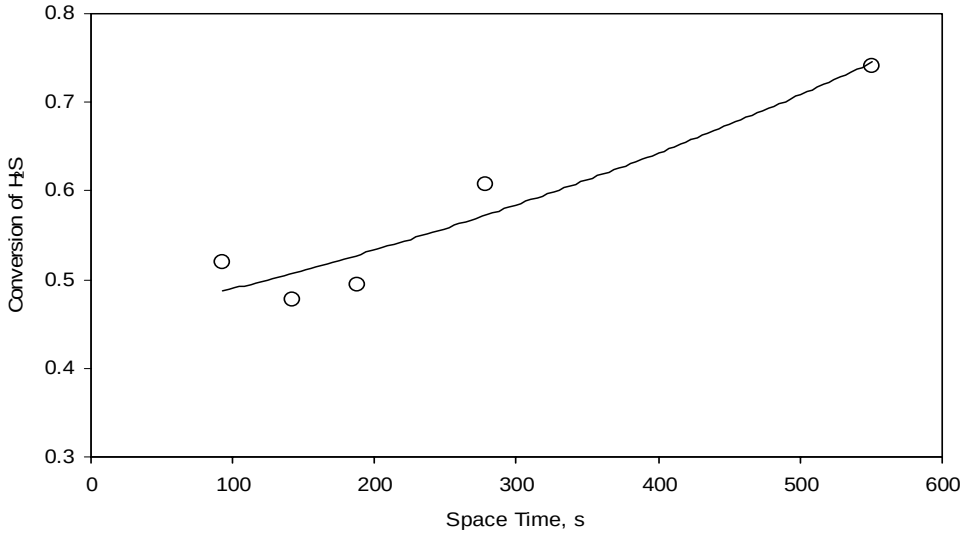
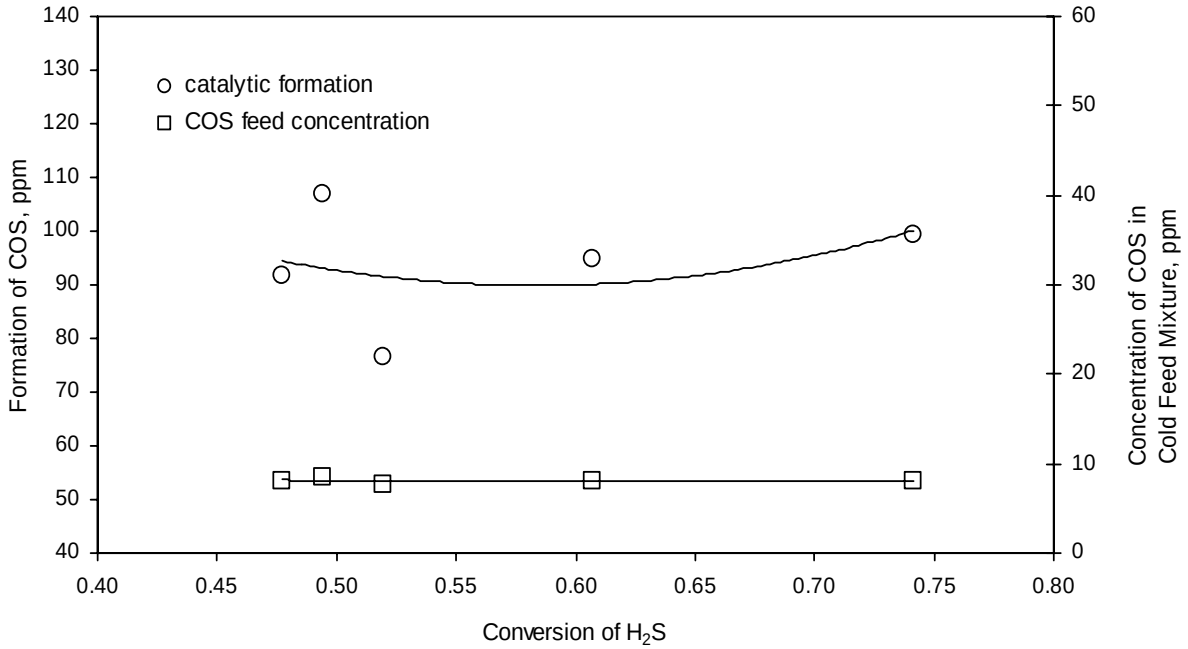


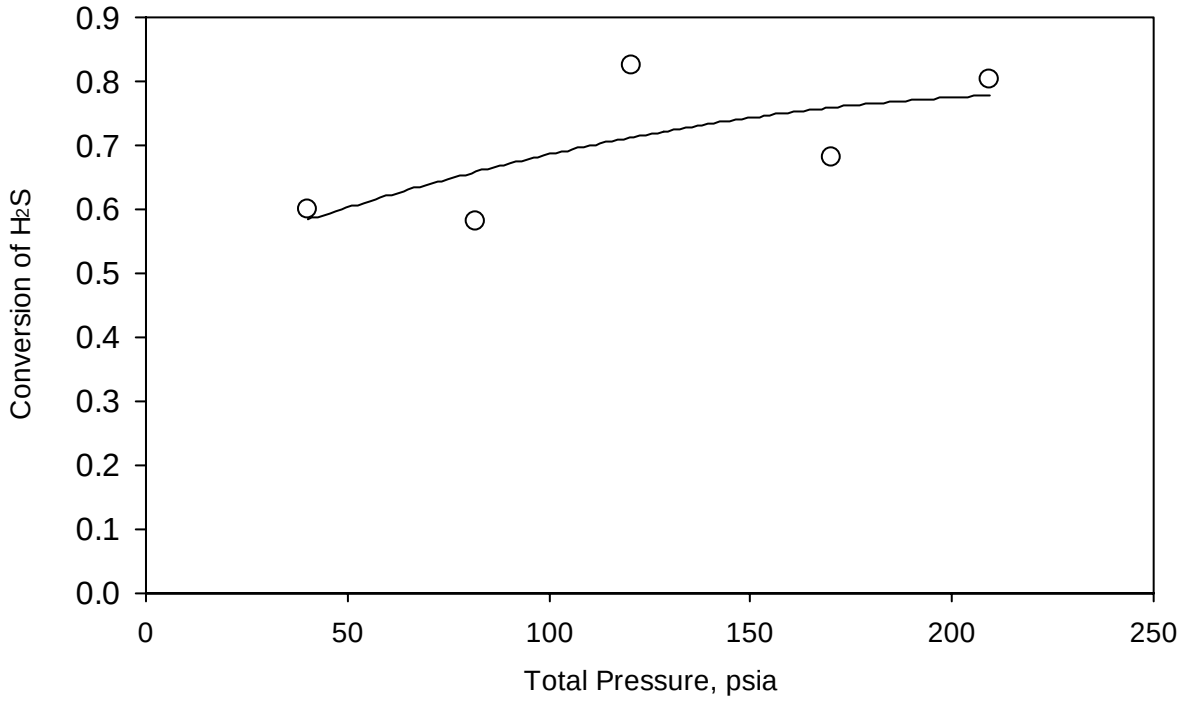
Figure 7. Effects of conversion of H_2S on formation of COS with 30 - 180 SCCM feed stream containing 3500-ppmv H_2S, 1800-ppmv SO_2, 37-v% CO, 10-v% CO_2, 24-v% H_2 and 10-v % moisture at 140°C, 117 -124 psia, and 90 - 570 s space time.



Effects of Total Pressure on Conversion of H_2S into Elemental Sulfur and formation of COS

Experiments on conversion of hydrogen sulfide to elemental sulfur and formation of COS with a 2-cm-diameter 15-cm-long γ-alumina wash-coated 400-cells/inch^2 monolithic catalyst were carried out over the space time range of 46 - 243 s to evaluate effects of total pressure on conversion of hydrogen sulfide to elemental sulfur and formation of COS at 140^{o}C and 40 – 210 psia. Gas mixtures are fed to a monolithic catalyst reactor containing 3,500 – 3600 ppmv H_2S, 1,800 ppmv SO_2, 37-v% CO, 24-v% hydrogen, 10-v% moisture, 10-v% CO_2, and nitrogen as remainder. Volumetric feed rates of gas mixtures to the monolithic catalyst reactor are 120 SCCM. Conversion of H_2S to elemental sulfur is 0.53 – 0.86.

Figure 8. Effects of total pressure on conversion of H_2S with a 120-SCCM feed stream containing 3500 - 3600 ppmv H_2S, 1800-ppmv SO_2, 37-v% CO, 10-v% CO_2, 24-v% H_2 and 10-v% moisture at 140^{o}C, 40 – 210 psia and 46 - 243 s space time.



Conversion of H_2S increases with increased total pressure, as shown in Figure 8. Catalytic formation of COS for reaction runs and thermal formation of COS for blank runs increase with increased total pressure. Catalytic formation of COS is not significantly different from thermal formation of COS at 140^{o}C and 40 – 210 psia, as shown in Figure 9. Catalytic formation of COS is independent of H_2S conversion over the H_2S conversion range of 0.68 – 0.83, as shown in Figure 10.

Figure 9. Effects of total pressure on formation of COS with a 120-SCCM feed stream containing 3500 - 3600 ppmv H_2S, 1800-ppmv SO_2, 37-v% CO, 10-v% CO_2, 24-v% H_2 and 10-v% moisture at 140°C, 40 -210 psia and 46 - 243 s space time.

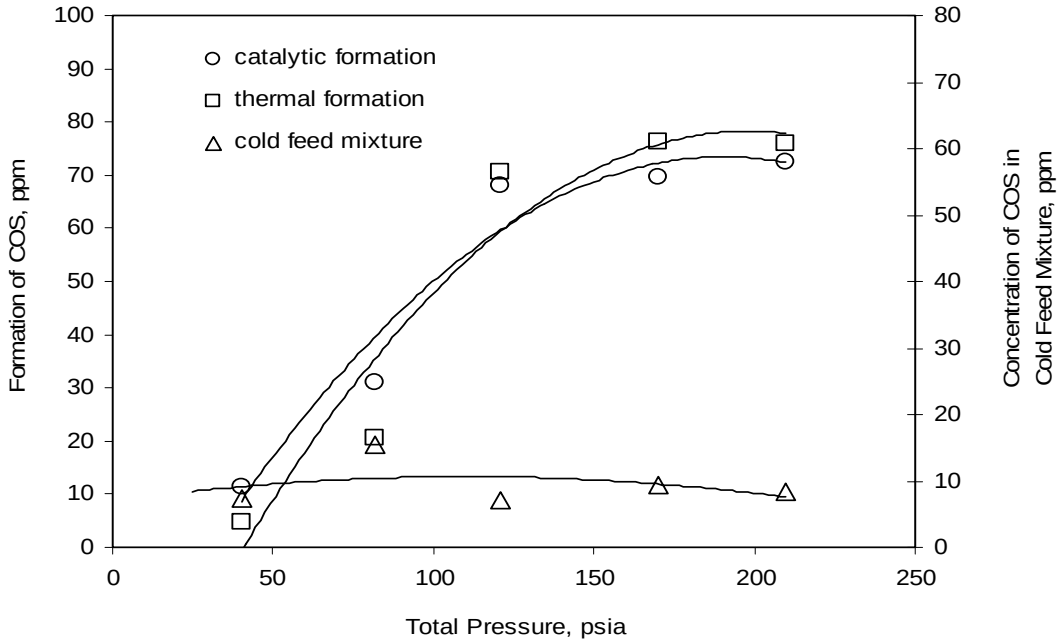
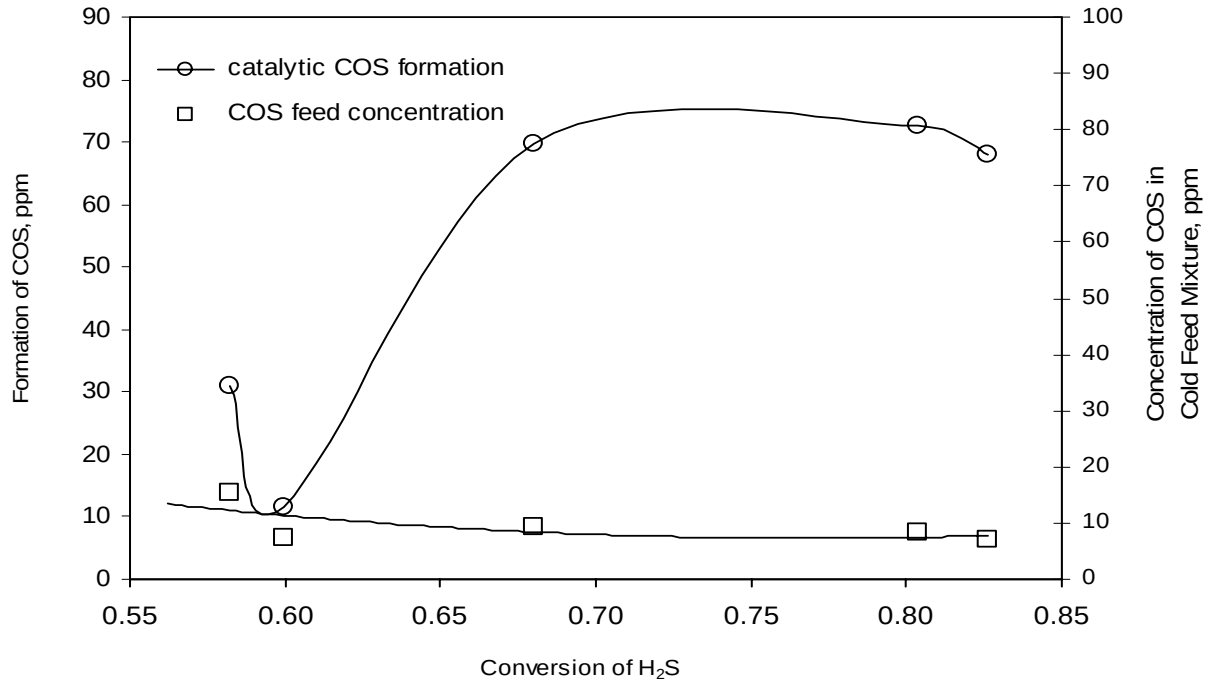


Figure 10. Effects of conversion of H_2S on formation of COS with a 120-SCCM feed stream containing 3500 - 3600 ppmv H_2S, 1800-ppmv SO_2, 37-v% CO, 10-v% CO_2, 24-v% H_2 and 10-v% moisture at 140°C, 40 - 210 psia and 46 - 243 s space time.



Regeneration of Catalyst for Conversion of H_2S into Elemental Sulfur and Formation of COS

Effects of catalyst age on conversion of H_2S to elemental sulfur and formation of COS were examined at 140°C, 119 -123 psia, and 138 – 142 s space time. The feed gas mixture to a monolithic catalyst reactor contains 3,600-ppmv H_2S, 1,800-ppmv SO_2, 37-v% CO, 24-v% H_2, 10-v% moisture, 10-v% CO_2, and N_2 as remainder. The volumetric feed rate of the feed gas mixture is 120 SCCM. The monolithic catalyst is regenerated with nitrogen at 140°C overnight. Conversion of H_2S to elemental sulfur does not decrease significantly with increased catalyst age up to 3900 hr catalyst age, as shown in Figure 11. Catalytic formation of COS decreases with increased catalyst age, as shown in Figure 12. Each reaction experimental run proceeds after a blank experimental run, which is carried out in the absence of moisture and a monolithic catalyst. Thermal formation of COS for the blank runs is lower than catalytic formation for the reaction runs, as shown in Figure 12. This observation may indicate that COS be formed in the presence of catalyst and moisture by reacting CO with elemental sulfur vapor, which is produced from the removal reaction of H_2S.

Figure 11. Effects of catalyst regeneration on conversion of H_2S to elemental sulfur with a 120-SCCM feed stream containing 3600-ppmv H_2S, 1800-ppmv SO_2, 37-v% CO, 10-v% CO_2, 24-v% H_2 and 10-v% moisture at 140°C, 119 - 123 psia and 138 – 142 s space time, regenerating the monolithic catalyst with N_2 overnight at 140°C.

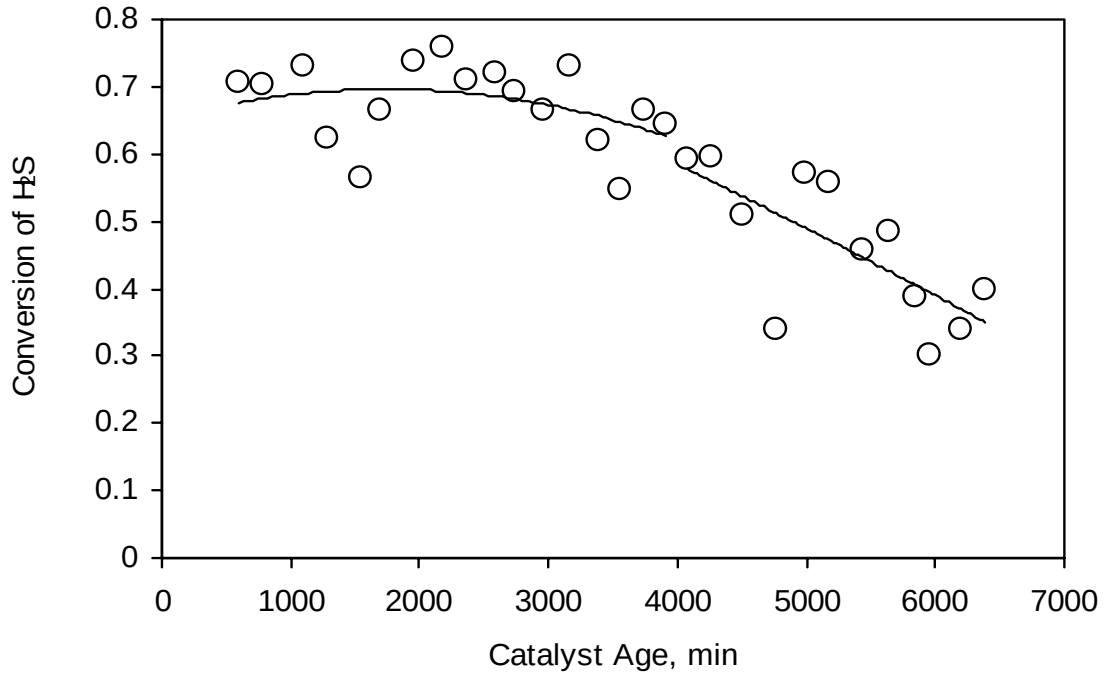
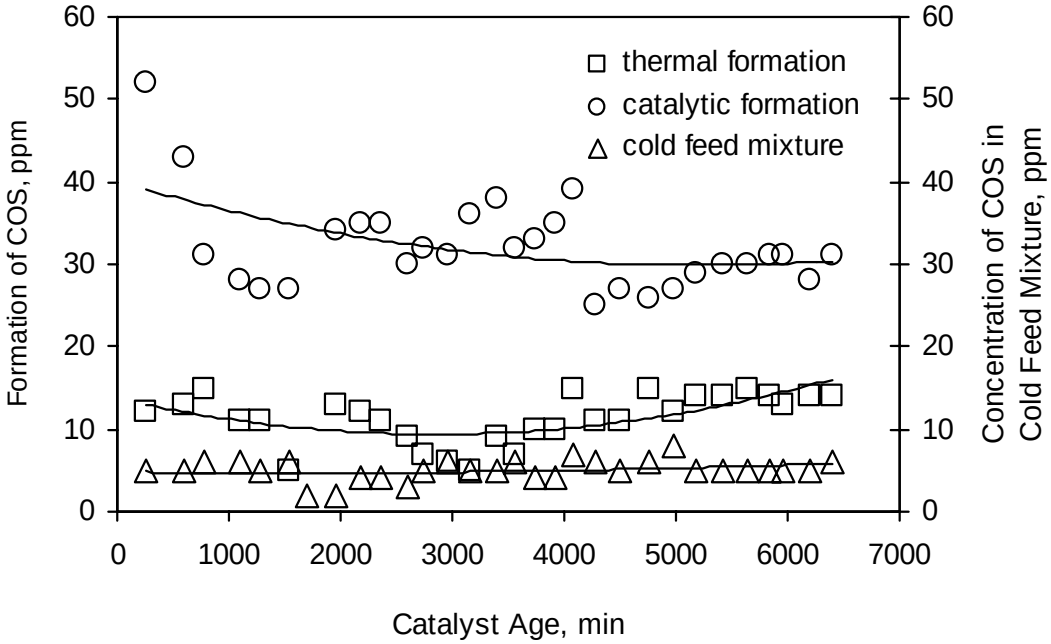


Figure 12. Effects of catalyst age on formation of COS with a 120 SCCM feed stream containing 3,600-ppmv H_2S, 1800-ppmv SO_2 as an oxidant, 37-v% CO, 10-v% CO_2, 24-v% H_2 and 10-v% moisture at 140°C, 119 -123 psia, and 138 – 142 s space time, regenerating the monolithic catalyst with N_2 overnight at 140°C.



CONCLUSIONS

The following conclusions were drawn based on experimental data generated from the monolithic catalyst reactor system, and their interpretations. Each reaction experimental run proceeds after a blank experimental run, which is carried out in the absence of moisture and a monolithic catalyst.

Conversion of H_2S to elemental sulfur does not follow the Arrhenius' equation. Conversion of H_2S to elemental sulfur increases with increased reaction temperature over the temperature range of 120 -140°C, whereas conversion of H_2S to elemental sulfur decreases with increased reaction temperature over the temperature range of 140 -150°C. Catalytic formation of COS levels off and is lowest over the temperature range of 120 -130°C, increases with increased reaction temperature over the temperature range of 130 -140°C, and decreases with increased reaction temperature over the temperature range of 140 -150°C. Thermal formation of COS is minimum over the temperature range of 120 -130°C, and increases with increased reaction temperature over the temperature range of 130 -150°C. Catalytic formation of COS is higher than thermal formation of COS. Catalytic formation of COS is minimum over the H_2S conversion range of 0.72 – 0.75 and also over the temperature range of 120 – 130°C.

Formation of COS for both the reaction runs and the blank runs increases with space time over the space time range of 90 – 190 s and decreases with increased space time over the space time range of 190 – 550 s. Catalytic formation of COS for the reaction runs is slightly lower than thermal formation of COS for the blank runs over the space time range of 140 – 280 s. Catalytic formation of COS for the reaction runs is much lower than thermal formation of COS for the blank runs at the space time 90 s, whereas catalytic formation of COS for the reaction runs is much higher than thermal formation of COS for the blank runs at the space time 550 s. Conversion of H_2S to elemental sulfur increases with space time. Catalytic formation of COS for the reaction runs appears to be independent of conversion of H_2S to elemental sulfur over the space time range of 90 – 570 s. These facts may suggest that COS be formed by reacting H_2S with CO ($CO + H_2S \rightarrow COS + H_2$)

Conversion of H_2S increases with increased total pressure. Catalytic formation for reaction runs of COS and thermal formation of COS for blank runs also increase with increased total pressure. Catalytic formation of COS is not significantly different from thermal formation of COS at 140^oC and 40 – 210 psia. Catalytic formation of COS is independent of H_2S conversion over the H_2S conversion range of 0.68 – 0.83.

Conversion of H_2S to elemental sulfur does not decrease significantly with increased age of 140^oC-nitrogen regenerated catalyst up to 3900 hr catalyst age. Catalytic formation of COS decreases with increased age of regenerated catalyst. Thermal formation of COS for the blank runs is lower than catalytic formation of COS for the reaction runs. This observation may suggest that COS is formed in the presence of catalyst and moisture by reacting CO with elemental sulfur vapor from the removal reaction of H_2S.

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